Phase transition and critical properties of spin-orbital interacting systems

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Phase transition and critical properties of Ising-like spin-orbital interacting systems in 2-dimensional triangular lattice are investigated. We first show that the ground state of the system is a composite spin-orbital ferro-ordered phase. Though Landau effective field theory predicts the second-order phase transition of the composite spin-orbital order, however, the critical exponents obtained by the renormalization group approach demonstrate that the spin-orbital order-disorder transition is far from the second-order, rather, it is more close to the first-order. The unusual critical behavior near the transition point is attributed to the fractionalization of the composite order parameter.

Keywords: spin-orbital systems; phase transition; critical exponents.

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I. INTRODUCTION

In many transition-metal-oxide insulators, in addition to spin degree of freedom, orbital degree of freedom plays important roles and leads to complicated phase diagram and interesting phenomena, such as orbital ordering (OO), metal-insulator transition (MIT) and colossal magnetoresistive effect, etc. In these insulating compounds, the sign and magnitude of the superexchange constants between magnetic ions depend on the regular orbital occupations, i.e. the OO[1,2,3]. Though there are a few of debates on the microscopic origin of OO, various experimental observations by the resonant Raman technique[4] and the resonant x-ray scattering (RXS)[5] have shown the existence of long-range OO in manganites and some other perovskite transition-metal oxides. The importance of the orbital degree of freedom also exhibits in new molecular compounds based on C_{60}[6], layered fullerides, and some two-dimensional (2D) copolymers[7].

The low-energy physics of these orbital insulators with both spin and orbital degrees of freedom is described by spin-orbital superexchange interactions. The interactions are usually highly anisotropic in orbital space, as well as in real space, due to the Hund's rule and the anisotropy of the orbital wavefunctions. Kugel and Khomskii[1] first proposed such an effective spin-orbital model to address the complicated magnetic structures. Such a theory incorporating the Jahn-Teller (JT) effect had witnessed a great success in past decades in explaining the magnetic structures of a wide range of material with the e_g orbital degeneracy, such as $KCuF_3$, $LaMnO_3$ and MnF_3, etc. However recent experiments in pseudocubic perovskite titanates[8], and vanadates[9] showed that in these insulators the static OO is absent and the orbital fluctuations and spin fluctuations should be taken into consideration on equal foots. Especially, the orbital degree of freedom entangled with spin degree of freedom may lead to the violation of the Goodenough-Kanamori rules[10] in these spin-orbital interacting systems, and the spin-orbital entanglement may be used to characterize different quantum phases and their competition[11] due to the composite spin-orbital fluctuations, indicating that the thermodynamic phase transition and critical properties are distinct from those of magnetic systems with only spin degree of freedom. The ground states and the low-energy excitations of these spin-orbital interacting insulators have been well understood through numerous theoretical and experimental efforts in the past decades. While, with the increasing temperature, the spin-orbital compounds exhibit complicated multicritical properties,

how the critical properties of the spin-orbital compounds behave near the phase transition point is seldom investigated. Further, it is important to understand the universal class of the spin-orbital model in statistical mechanics and condensed matter theory.

In this Letter we present the phase transition and critical properties of an Ising-like spin-orbital system, where the long-range order and the order parameter of the system are composite spin and orbital order, rather than single spin or orbital order. We show that due to such an entanglement, the critical exponents of the spin-orbital interacting systems are unexpectedly different from those in magnetic systems without orbital degeneracy. To demonstrate the general characters of the Ising-like spin-orbital model, we first determine the spin-orbital ordered ground state utilizing the cluster self-consistent field (Cluster-SCF) approach, and then obtain the evolution of the spin-orbital order parameter with temperature in the Landau mean-field approximation. Further, the non-trivial critical exponents, which characterize the power-law behaviors of the order parameter of the system, are also obtained by Wilson's renormalization group (RG) technique. And in the final we briefly discuss our results.

II. MODEL HAMILTONIAN AND GROUND STATE

The effective superexchange interaction describing the low-energy processes in orbital-degenerate transition-metal oxides takes the generalized Heisenberg form[1]

$$H = -\sum_{<i,j>} [J_S \mathbf{S}_i \cdot \mathbf{S}_j + f(\mathbf{T}_i, \mathbf{T}_j) + g(\mathbf{T}_i, \mathbf{T}_j)\mathbf{S}_i \cdot \mathbf{S}_j] \tag{1}$$

where J_S , $f(\mathbf{T}_i, \mathbf{T}_j)$ and $g(\mathbf{T}_i, \mathbf{T}_j)$ represent the spin, orbital and spin-orbital superexchange constants, respectively, which originate from the quantum mechanical intermediate virtual processes in the strong correlation regime. The Hamiltonian (1) describes the spin-spin, orbital-orbital and spin-orbital couplings with the $SU(2)$ symmetry in spin space in the system with S=1/2 and T=1/2. In the realistic compounds, the spin $SU(2)$ symmetry of the systems is usually broken by the magnetic anisotropy. Moreover, the symmetry of the orbital space is broken by the Hund's coupling and the anisotropy of the hopping integrals $t_{\alpha\beta}$, which depends on the shapes of the 3d-orbital wave functions and the relative orientations of two nearest-neighbor d-orbits. Thus the effective spin-orbital interactions in realistic compounds are highly anisotropic.

The simplest theoretical model describing the anisotropic spin-orbital interactions in an insulator reads

$$H = -\sum_{<ij>} JS_i^z T_i^z \cdot S_j^z T_j^z + h\sum_i S_i^z T_i^z \tag{2}$$

where the operator S_i^z labels the z-component of the electron spin, while T_i^z denote the z-component of the orbital pseudo-spin opertaor. In what follows we let S=$\pm$1 and T=$\pm$1. The summation in the first term is taken over the nearest-neighbor pairs $< ij >$; h stands for the conjugated field of the spin-orbital order parameter. Such an ideal model, like the Ising model for ferromagnetism, can serve as important reference in understanding the essential characters of the phase transition in the spin-orbital interacting systems. In what follows, we consider the spin-orbital system in a 2D triangular lattice, since more and more transition-metal oxides, such as $LiVO_2$[12] and Na_xNiO_2[13], are found to be triangular lattice.

To get a straight insight into the physical properties of strongly correlated spin-orbital systems in 2D triangular lattice, we utilize the cluster self-consistent field (SCF) approach developed recently[14] to extract the ground state properties of the Ising-like spin-orbital model (2) at T=0 K. The numerical results showed that in the 3-site cluster, the groundstate energy per site is $E_g = -0.5625J$, while the macroscopic groundstate degeneracy is not completely removed at $h = 0$. Such a degeneracy is removed and the ground state is ferro-ordering in the limit of $h \to 0$. As a comparison, the ground state of the spin Ising model is non-degenerate ferromagnetic at $h = 0$. Nevertheless, the spin-orbital correlation functions between sites in the cluster are ferro-ordering, while the spin-spin and the orbital-orbital correlation functions vanish, suggesting that the spin and the orbital degrees of freedom form an entangled triplet, and lead to the composite ferro-order, similar to the spin-orbital ferromagnetic order found by Zhang and Shen[15] in the $SU(4)$ spin-orbital model in 2D square lattice. We notice that the degeneracy of each site in the present model is fourfold, and the ground state is degenerate "ferromagnetic", this is comparable with the Potts model with $p = 4$, whose critical behavior had been widely studied by Baxter *et al.*[16] and Nienhuis *et al.*[17], and the ground state forms a p-fold degenerate ferromagnetic state, corresponding to a uniform classical state. However, the ground state of the present mdoel is spin-orbital entangled with $< ST >$=1.

III. MEAN-FIELD THEORY

With increasing temperature until to a critical value T_c, the composite spin-orbital order is stable against the thermal fluctuation. We adopt the Bethe approximation[18] to study the temperature evolution of the order parameter. Considering a 7-site cluster consisting of a central site with 6 nearest neighbours. The intra-couplings between the central site and 6 nearest neighbours are accurately calculated, while the inter-couplings between the nearest-neighbour sites and other lattice sites are treated as a mean field h^{MF}. In the Bethe approximation[18], we find that in agreement with the preceding results, $< S^z >= 0$, and $< T^z >= 0$, however, the average of the combination of spin and orbit, $< S^z T^z >$, is finite and given by the self-consistent equation:

$$< S^z T^z >= (z-1) Ln \frac{1+e^{2\alpha+2\gamma}}{e^{2\alpha}+e^{2\gamma}} \tag{3}$$

in the absence of external field h, here $\alpha = J < S^z T^z > /k_B T$ and $\gamma = J/k_B T$. Moreover, Eq.(3) gives rise to a finite critical temperature at $T_C = \frac{J}{k_B}/8\ln(z/(z-2)) \simeq 0.303\frac{J}{k_B}$. In fact, due to spin-orbital entanglement, the Hamiltonian (2) can be rewritten as complete Ising-like:

$$H = -J\sum_{<ij>} Q_i Q_j - h\sum_i Q_i$$

with the composite operator $Q_i = S_i^z T_i^z$. Thus one would expect that the phase transition and critical behavior of the present spin-orbital interacting system quite resembles to that of the spin Ising model. For example, one expects that the order-disorder transition of the present system is the second order.

To further illustrate the evolution of the composite order parameter with temperature near the critical region, we consider the high-temperature expansion of the free energy in the *Landau* mean field (MF) approximation. The order parameter, $M =< S^z T^z >$, is a small quantity in this situation. The *Landau* free energy per site becomes

$$\begin{aligned} F/N &= -2k_B T \ln 2 + M^2 J + \ln(\cosh\frac{\beta z M}{2}) \\ &\simeq -2k_B T \ln 2 + (zJ - \frac{\beta z^2}{8})M^2 + \frac{\beta^3 z^4}{192}M^4 \end{aligned} \tag{4}$$

where N is the total number of lattice sites, and z=6 is the coordinate number. Minimizing F with respect to M gives rise to the mean-field critical behavior of the composite spin-orbital ferro-order : $M \propto (T_c - T)^{1/2}$, near the critical point, and the corresponding critical

temperature is given by $T_C = zJ/8k_B \simeq 0.375J/k_B$, a slightly large than T_C obtained by the Bethe approximation.

IV. RENORMALIZATION GROUP APPROACH

Obviously, the present Curie point is overestimated in comparison with that obtained by Bethe approximation. On the other hand, the present *Landau* mean-field approximation becomes unreliable, it underestimates the thermal fluctuations near the critical regime, and gives incorrect critical exponents near the transition point. It is well known that the universal characters and the scaling laws of the thermal quantities are not exactly captured by the mean-field theory. which appeals for the renormalization group calculations for the model (2).

In the real-space renomalization group, the triangular lattice is divided into the 3-site cells, which interact with each other in the similar ways as the original one as shown in Fig.1. The transformation procedure to the spin-orbital interactions (2) is implemented by the RG technique proposed by Niemeijer and van Leeuwen[19]. In the present cell structure in Fig. 1, we define the cell spin and the cell orbit as

$$S_i^{'} = sign(\sum_{i=1}^{x} S_i); \qquad T_i^{'} = sign(\sum_{i=1}^{x} T_i) \tag{5}$$

A cell contains 16 different internal configurations which can be labeled by the direct product of the spin sub-configurations $\{\sigma\}$ and the orbital sub-configurations $\{\tau\}$. By expressing the S_i in terms of $S_i^{'}$ and σ and the T_i in terms of $T_i^{'}$ and τ, the transformation of Hamiltonian between the cell system and the site system is defined as,

$$exp(H^{'}(S_i^{'}, T_i^{'})) = \sum_{\{\sigma\}} \sum_{\{\tau\}} exp(H(S_i^{'}\sigma; T_i^{'}\tau)) \tag{6}$$

with $\{\sigma\}$ and $\{\tau\}$ running over all internal configurations; here the reverse temperature factor $-\beta$ is absorbed in the coupling constants.

To approximately calculate the renormalization equation (6), we utilize the cumulant expansion method[19]. Spliting the Hamiltonian (6) into an internal part H^0 and a remainder part V and treating the latter as a perturbation, one has the cell Hamiltonian in the second-

order expansion of V,

$$
\begin{aligned}
&H^{'}(S_i^{'}, T_i^{'}) \simeq \ln < exp(V) > \\
&\simeq < V >_0 + \frac{1}{2}(< V^2 >_0 - < V >_0^2) + O(V^3)
\end{aligned}
\tag{7}
$$

where

$$
< A >_0 = \frac{\sum_{\{\sigma\}} \sum_{\{\tau\}} A(\sigma,\tau) exp(H^0(\sigma,\tau))}{\sum_{\{\sigma\}} \sum_{\{\tau\}} exp(H^0(\sigma,\tau))}.
$$

In the absence of the conjugated field h, the evaluation to (6) is straightforward, and leads to a set of renormalization equations:

$$
\begin{aligned}
K^{'} &= 2f_1^2 K + 8K^2 f_1^2 (1 + f_2 - 2f_1^2) + 3f_1^2 L + 2f_1^2 M \\
L^{'} &= 2(1 + 7f_2 - 8f_1^2) f_1^2 K^2 + f_1^2 M \\
M^{'} &= 8(f_2 - f_1^2) f_1^2 K^2
\end{aligned}
\tag{8}
$$

with

$$
\begin{aligned}
f_1 &= \exp 3K/(\exp 3K + 3\exp -K) \\
f_2 &= (\exp 3K - \exp -K)/(\exp 3K + 3\exp -K).
\end{aligned}
$$

The numerical results for the nontrivial fixed points of the nonlinear equations (8) are shown in *Table I*. For comparison, the results of the Ising model with $S = \frac{1}{2}$[19] and 1 and XY model[20] are also presented. The comparisons of the critical exponents for various models are shown in *Table II* and *III*, respectively. The presence of the unstable fixed points suggests that the existence of the spin-orbital order-disorder phase transition.

Linearizing the renormalization equations (8), and diagonalizing the matrix, $T_{\alpha\beta} = \partial K_\alpha / \partial K_\beta$, around the fixed points, we get only one relevant eigenvalue, λ_T, in the second-order cumulant expansion, as shown in *Table II*. In the presence of the conjugated field h, analogous to Ref.[19], we have yielded the magnetic relevant eigenvalue,

$$
\lambda_H = \frac{\partial h'}{\partial h} = 3f_1 + 12 f_1 K (1 + 2f_2 - 3f_1^2) \tag{9}
$$

in the first-order cumulant expansion, which is also listed in *Table III*. Two corresponding "thermal" and "magnetic" exponents, obtained through $\alpha_{T(h)} = d \ln L / d \ln \lambda_{T(h)}$, are also listed in Table II and III, respectively. Thus, according to the scaling laws, the critical

exponents of the specific heat, α, with respect to $T - T_C$ and of the order parameter, δ, with respect to the conjugated field at $T = T_C$ are also known, and listed in *Table.II* and *III*, respectively. Moreover, the critical temperature can be evaluated as follows

$$K_C(J) = K_C^* - \sum_{\alpha \neq n.n} (\varphi_\alpha^T / \varphi_{n.n}^T) K_\alpha^* \tag{10}$$

where φ_α is the left eigenvectors of the matrix $T_{\alpha\beta}$. The result is also listed in *Table II*.

From *Table II*, one finds that the critical exponent of specific heat α is far from that of the Ising model with the second-order phase transition, and the 'thermal' exponent α_T lies between the second-order and the first order, and α_T is more close to the first order, implying that the present spin-orbital order-disorder phase transition of the composite order parameter, $< S^z T^z >$, is of a weak second order in the absence of the conjugated field. Such a weak second-order phase transition can be clearly seen by rewriting the singular free energy near the critical point[19],

$$f_{sing} = A\mu_1^{\alpha_{T(H)}} + O(\mu_1) \tag{11}$$

where μ_1 represents the distance from the fixed points. Thus the order of the singularity of the free energy is solely determined by the 'thermal' ('magnetic') exponent $\alpha_{T(H)}$. One finds that the 'thermal' exponents are close to but larger than unity, which are different from those of the second-order transition in the spin-$\frac{1}{2}$ and the spin-1 Ising models, as seen in *Table II* and *III*. Meanwhile, magnetic eigenvalue α_H, which is close to but smaller than unity, suggests that the phase transition induced by the conjugated field is weak first-order.

Interestingly, we note that in *Table II* and *III* the present critical exponents of the spin-orbital model are close to those of the two-dimensional quantum XY model[20], suggesting that these two models belong to the same universal class. Since the component of order parameter of the quantum XY model is two-dimensional, this implies that near the critical point, the order parameter of the present spin-orbital model decouples into two independent components, i.e., the spin and the orbital degrees of freedom have already disentangled. This result is significantly different from the prediction of the mean-field theory. Moreover, the critical temperature obtained by the RG approach is considerably reduced, in comparison with the mean-field result, indicating that the strong fluctuations near critical points are considered properly. We also notice that the critical temperature is much smaller than that of the spin Ising model. Therefore, we conclude that the thermal fluctuations are expectedly strong, due to the disentanglement between spin and orbital degrees of freedom.

V. REMARKS AND SUMMARY

As we state in the preceding, the formation of the spin-orbital composite order parameter arises from the entanglement of the spin and the orbital degrees of freedom at the same site. The entanglement between the spin and orbit disappears when the system is close enough to the critical point, and strong thermal fluctuations entirely destruct the composite spin-orbital order. The considerable discrepancy of the critical exponents between the RG approach and the *Landau* mean-field theory arises from the fractionalization of the composite order parameter due to the disentanglement between the spin and orbital degree of freedom at critical point, which was not captured by the mean-field theory. More interestingly, the fractionalization of the order parameter is quiet similar to the deconfinement critical phenomena[21].

As well known, the orbital phenomena in most of spin-orbital compounds are relevant to lattice distortion, which is generally believed to cause the first-order phase transition in many transition-metal oxides. The present results show that the essential of the phase transition in spin-orbital interacting system deviates from the second-order, and more close to the first order. We expect more experiments to testify our prediction. Also, we realize that the present negative value for δ can be attributed to that we just consider the first-order and the second-order approximation in cumulant expansion approach, thus it is expected that including higher order approximation[22] will refine and provide more accurate critical exponents.

In conclusion, we have obtained the critical exponents of the phase transition of the Ising-like spin-orbital model, and shown that the order-disorder phase transition behaves as a weakly first-order; and due to fractionalization of the composite order parameter, the spin-orbital system may belong to the same universal class as the XY model.

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FIGURE CAPTION

Fig.1 Sketched 3-site cells on the triangular lattice, see the shaded triangles.

TABLE CAPTIONS

Table I Fixed points of the renormalization equations of Ising models with $S = 1/2$[19] and 1, present model (SO), and XY model[20].

Table II Thermal eigenvalue λ_T, 'thermal' exponent α_T, critical temperature K_C and critical exponent α of Ising models with $S = \frac{1}{2}$[19] and 1, present model (SO), and XY model[20].

Table III Magnetic eigenvalue λ_H, 'magnetic' exponent α_H and critical exponent δ of Ising models with $S = 1/2$[19] and 1, present model (SO), and XY model[20].

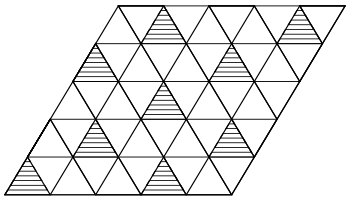

FIG. 1:

TABLE I:

Model		Ising(s=1/2)	Ising(s=1)	S-O	XY
first-order	K^*	0.3356	0.6312	0.4950	nonpoint
second-order	K^*	0.3350	0.4791	0.2568	0.8554
	L^*	-0.014	-0.011	0.0422	0.2131
	N^*	-0.015	-0.019	0.0095	-0.203

TABLE II:

Model	λ_T	α_T	K_C	α
Ising(s=1/2)	1.7835	1.8988	0.2514	0.1012
Ising(s=1)	1.7909	1.8853	0.4792	0.1147
S-O	2.4499	1.2260	0.2669	0.7740
XY Model	2.5410	1.1780	0.8665	0.8015

TABLE III:

Model	Ising(s=1/2)	Ising(s=1)	S-O	XY
λ_H	3.0570	3.2838	5.1410	4.9600
α_H	0.9831	0.9240	0.6710	0.6904
δ	-59.17	-13.16	-3.039	-3.230